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(54) Title of the Invention: Azeotropic or azeotrope-like composition
comprising fluorine containing acetal and butanol

(57) Abstract

20 Problem:

To provide novel azeotropic compositions and azeotrope-like
compositions with which there is no loss of the excellent properties such as
the cleaning properties and the low toxicity, for example, of the fluorinated
hydrocarbons which contain chlorine and the chlorinated hydrocarbons,
25 with which there is no concern with respect to the destruction of the ozone
layer and which have little global warming effect.

Means of Resolution:

Azeotropic compositions or azeotrope-like compositions which
contain 1,1,1,3,3,3-hexafluoro-2-[(2,2,2-trifluoro-1-
30 trifluoromethylethoxy)methoxy]propane which can be represented by the
formula $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ and at least one type of alcohol
selected from among 1-butanol and 2-butanol.

Scope of the Patent Claims

[Claim 1]

An azeotropic composition or azeotrope-like composition which contains 1,1,1,3,3,3-hexafluoro-2-[(2,2,2-trifluoro-1-trifluoromethylethoxy)methoxy]propane and at least one type of alcohol selected from among 1-butanol and 2-butanol.

Detailed Description of the Invention

[0001]

Technical Field of the Invention

The invention concerns azeotropic compositions or azeotrope-like compositions, and more precisely the invention concerns azeotropic compositions or azeotrope-like compositions which can be used for flux cleaning, degreasing cleaning, water-cutting drying and as solvents.

[0002]

15

Prior Art

Halogenated hydrocarbons have long been very well known as cleaning solvents for electronic parts, precision machine parts and resin processing parts for example. These are characterized as a group of hydrocarbons which are substituted with chlorine atoms or with chlorine atoms and fluorine atoms. These halogenated hydrocarbons have low toxicity and become non-flammable as the number of substituted halogen atoms is increased and they are also both chemically and thermally stable and, moreover, they do not penetrate into the surfaces of plastics and rubbers and provide a level of solubility which is ideal for dissolving waxes and oils and fats and so they are widely used in a variety of industrial fields. For example, the known halogenated hydrocarbons include chlorinated hydrocarbons such as trichloroethylene, tetrachloroethylene and 1,1,1-trichloroethane, and freons such as 1,1,2-trichloro-1,2,2-trifluoroethane (freon 113), and the latter freon hydrocarbons in particular have low toxicity and are non-flammable, and both chemically and thermally stable and so they are used in a wide range of fields. However, it has been pointed out that since these fluorinated hydrocarbons which contain chlorine atoms and 1,1,1-trichloroethane do contain chlorine atoms

they have a serious disadvantage in that they destroy the ozone layer in the stratosphere and it has been decided internationally to stop the production and use of these materials. Furthermore, trichloroethylene and tetrachloroethylene have resulted in problems with ground water contamination and as a result of subsequent investigation of the state of environmental pollution and as a result of an investigation into their toxicity such as their chronic toxicity it has been ordered that these be designated as class 2 specified chemicals from 1989. In view of these circumstances the development of materials to replace these fluorinated hydrocarbons which contain chlorine and chlorinated hydrocarbons has been carried out actively. At the present time fluorinated compounds which do not contain chlorine atoms are well known as substitute materials. However, compounds of this type lack dissolving power for waxes and oils and fats since they do not contain chlorine atoms. In order to resolve this problem general purpose solvents such as alcohols have been mixed with the substitute compounds with a view to improving the dissolving power, but when mixtures of this type are used with heating as cleaning agent it is known that the liquid composition and the vapour composition are different so that concentration control is required. The azeotropic composition in a mixed composition is such that the vapour composition and the liquid composition are the same, and an azeotrope-like composition is such that they are almost the same, and these are known as useful compositions where concentration control is not required (Japanese Unexamined Patent Application Laid Open H8-268944). However, a mixture does not always have an azeotropic composition, and whether it will or will not have such a composition cannot be predicted.

[0003]

Problems to be Resolved by the Invention

The problem for the present invention is to provide a novel azeotropic compositions and azeotrope-like compositions with which there is no loss of the excellent properties such as the cleaning properties and the low toxicity, for example, of the fluorinated hydrocarbons which contain chlorine and the chlorinated hydrocarbons, with which there is no concern

Means of Resolving These Problems

[0005]

15 Compositions such as those indicated below are included in practice among the compositions of this invention. Moreover, an azeotropic composition signifies a composition of which the vapour composition and the liquid composition are the same and with which there is no change in the composition of the composition after repeated
20 vaporization and condensation. Furthermore, an azeotrope-like composition signifies a composition of which the vapour composition and the liquid composition are almost the same and with which there is only a change of an order which can be neglected in the composition of the composition after repeated vaporization and condensation.

[0006]

(2) The azeotrope-like compositions comprise from 30.0 to 60.0 wt% of $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ and from 40.0 to 70.0 wt% of 2-butanol, and the preferred azeotropic composition comprises 48.36 wt% of $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ and 51.64 wt% of 2-butanol. The boiling point of this azeotropic composition is 97.87°C at atmospheric pressure (760 mmHg).

[0007]

Moreover, the 1,1,1,3,3,3-hexafluoro-2-[(2,2,2-trifluoro-1-trifluoromethylethoxy)methoxy]propane which is used in the invention is a known substance and it can be obtained easily by reacting 1,1,1,3,3,3-hexafluoropropane-2-ol with 1,3,5-trioxane in the presence of a high concentration of sulphuric acid catalyst for example. Furthermore, in oral toxicity experiments with rats, $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ had a 50% lethal dose rate (LD_{50}) value higher than 2000 mg/kg and it gave a negative result in a mutagenicity test (Ames test) and it is clearly a compound of low toxicity. Moreover, on measuring the flash point of $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ in an enclosed system ignition was not seen to occur and it is a useful substance which can be employed effectively as a solvent.

[0008]

Various stabilizers may be added to the compositions of this invention when they are to be used under harsh conditions. Stabilizers which are distilled on distillation or which form an azeotrope-like mixture are preferred. Actual examples of such stabilizers include aliphatic nitro compounds such as nitromethane and nitroethane, aromatic nitro compounds such as nitrobenzene and nitrostyrene, ethers such as dimethoxymethane, 1,2-dimethoxyethane, 1,4-dioxane, 1,3,5-trioxane, epoxides such as glycidol, methyl glycidyl ether, allyl glycidyl ether, phenyl glycidyl ether, 1,2-butylene oxide, cyclohexene oxide and epichlorohydrin, unsaturated hydrocarbons such as hexene, heptene, pentadiene, cyclopentene and cyclohexene, olefinic alcohols such as allyl alcohol and 1-butene-3-ol, acetylene-based alcohols such as 3-methyl-1-butyne-3-ol and 3-methyl-1-pentyne-3-ol, and acrylic acid esters such as methyl acrylate,

ethyl acrylate, butyl acrylate and vinyl methacrylate. Moreover, phenols, amines and benzotriazoles can be used conjointly in order to obtain a synergistic stabilizing effect. These stabilizers may be used individually, or two or more types may be used conjointly. The amount of stabilizer used
5 differs according to the type of stabilizer, but it is of an extent such that the azeotrope-like properties are not affected adversely. The amount used is generally of the order of from 0.01 to 10 wt%, and preferably of the order of from 0.1 to 5 wt%, of the composition of this invention.

[0009]

10 Furthermore, various surfactants can be added to a composition of this invention, as required, in order to further improve the cleaning power and surface action etc. Examples of such surfactants include non-ionic surfactants such as sorbitan fatty acid esters, for example sorbitan mono-oleate and sorbitan trioleate, polyoxyethylene-sorbitan fatty acid esters, for
15 example polyoxyethylene sorbitan tetra-oleate, polyethylene glycol fatty acid esters such as polyoxyethylene monolaurate, polyoxyethylene alkyl ethers, for example polyoxyethylene lauryl ether, polyoxyethylene alkylphenyl ethers, for example polyoxyethylene nonylphenyl ether, and polyoxyethylene alkylamine fatty acid amides for example polyoxyethylene
20 oleic acid amide, and these may be used individually or a combination of two or more types can be used. Cationic surfactants or anionic surfactants can be used conjointly with these non-ionic surfactants with a view to improving the cleaning power and surface action synergistically. The amount of surfactant used differs according to the type, but it is of an
25 order which does not interfere with the azeotrope-like properties, and it is generally of the order of from 0.1 to 20 wt%, and preferably of the order of from 0.3 to 5 wt%, of the composition of this invention.

[0010]

30 The compositions of this invention can be used in a wide range of known cleaning and drying applications, and in particular they can be used as flux cleaning agents, cleaning solvents, degreasing cleaning agents and water-cutting drying agents and they are very useful as substitutes for the conventional freon 113 and 1,1,1-trichloroethane. Actual examples of such

applications include agents for the removal of flux, grease, oil, wax or ink for example, and cleaning agents and water-cutting agents for use with electronic parts (printed circuit boards, liquid crystal displays, magnetic recording parts, semiconductor materials and the like), electrical machine parts, precision machine parts, resin processing parts, optical lenses and clothing. Methods of cleaning which can be adopted include immersion, spraying, boiling, ultrasonic cleaning and vapour cleaning or a combination of such methods of cleaning. Furthermore, the compositions of this invention can be used in the same ways as in the past in various applications such as solvents for paints, extracting agents, heating media and foaming agents for example.

[0011]

Illustrative Examples

The invention is described in more detail below by means of illustrative examples.

[0012]

Example 1

Measurement of the Azeotrope Point of $(CF_3)_2CHOCH_2OCH(CF_3)_2$ and 1-butanol

The vapour/liquid equilibrium and azeotrope point of 1,1,1,3,3,3-hexafluoro-2-[(2,2,2-trifluoro-1-trifluoromethylethoxy)methoxy]propane which can be represented by the formula $(CF_3)_2CHOCH_2OCH(CF_3)_2$ and 1-butanol were measured using vapour/liquid equilibrium measuring apparatus. A mixed sample of a given composition of $(CF_3)_2CHOCH_2OCH(CF_3)_2$ and 1-butanol was introduced into the sample container and heated. The heating was adjusted in such a way as to provide an appropriate dripping rate for the vapour phase condensate and stable boiling was maintained for at least 40 minutes. After confirming that they were stable, the pressure and the boiling point were measured. The liquid phase and the vapour phase were sampled and composition analysis of the liquid samples was carried out using gas chromatography. The results of these measurements are shown in Table 1 and Figures 1 and 2. According to the results the compositions of this invention which contain

(CF₃)₂CHOCH₂OCH(CF₃)₂ in an amount ranging from 70.0 to 99.9 wt% and 1-butanol in an amount ranging from 0.1 to 30.0 wt% are azeotrope-like compositions. The composition comprising 79.39 wt% (CF₃)₂CHOCH₂OCH(CF₃)₂ and 20.61 wt% 1-butanol is the azeotropic composition, and the boiling point at atmospheric pressure (760 mmHg) is 108.89°C.

[0013]

Table 1

(CF₃)₂CHOCH₂OCH(CF₃)₂ + 1-Butanol System

Vapour/liquid equilibrium measurement results at 760 mmHg.

	t (°C)	x ₁ (wt%)	y ₁ (wt%)
1	117.66	0.00	0.00
2	114.42	17.08	43.28
3	112.56	30.03	56.39
4	110.96	43.62	65.69
5	109.76	59.54	72.19
6	109.11	70.41	76.10
7	108.92	79.37	79.38
8	108.89	79.69	79.52
9	108.97	81.50	80.38
10	109.24	86.16	82.90
11	109.66	89.14	85.02
12	110.40	92.10	87.59
13	111.56	94.43	90.37
14	113.87	97.21	94.28
15	116.43	98.88	97.55
16	118.82	100.00	100.00

x₁: Concentration of (CF₃)₂CHOCH₂OCH(CF₃)₂ in the liquid phase

y₁: Concentration of (CF₃)₂CHOCH₂OCH(CF₃)₂ in the vapour phase

15 [0014]

Example 2

Measurement of the Azeotrope Point of (CF₃)₂CHOCH₂OCH(CF₃)₂ and 2-butanol

The same procedure as in Example 1 was followed except that 2-butanol was used instead of 1-butanol. The results obtained are shown in

20

Table 2 and Figures 3 and 4. According to the results the compositions of this invention which contain $(CF_3)_2CHOCH_2OCH(CF_3)_2$ in an amount ranging from 30.0 to 60.0 wt% and 2-butanol in an amount ranging from 40.0 to 70.0 wt% are azeotrope-like compositions. The composition comprising 48.36 wt% $(CF_3)_2CHOCH_2OCH(CF_3)_2$ and 51.64 wt% 2-butanol is the azeotropic composition, and the boiling point (at 760 mmHg) in this case is 97.87°C.

[0015]

Table 2

$(CF_3)_2CHOCH_2OCH(CF_3)_2$ + 2-Butanol System

Vapour/liquid equilibrium measurement results at 760 mmHg

	t (°C)	x ₁ (wt%)	y ₁ (wt%)
1	99.48	0.00	0.00
2	98.47	20.17	27.78
3	98.12	31.04	37.02
4	97.92	41.43	44.24
5	97.87	46.83	47.44
6	97.87	48.61	48.53
7	97.87	50.29	49.52
8	97.87	51.20	50.01
9	97.83	51.44	50.18
10	97.97	54.75	52.12
11	98.03	58.90	54.39
12	98.20	64.67	57.68
13	98.57	70.56	61.41
14	99.06	76.03	65.14
15	99.73	80.33	68.38
16	101.18	85.87	73.56
17	103.49	89.87	79.28
18	105.78	93.02	84.02
19	109.56	95.93	89.71
20	113.79	98.26	95.11
21	118.82	100.00	100.00

x₁: Concentration of $(CF_3)_2CHOCH_2OCH(CF_3)_2$ in the liquid phase

y₁: Concentration of $(CF_3)_2CHOCH_2OCH(CF_3)_2$ in the vapour phase

[0016]

Reference Example 1

Synthesis of $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$

111.51 g (1.238 mol) of 1,3,5-Trioxane (molecular weight: 90.08)
5 and 1861.9 g (11.08 mol) of 1,1,1,3,3,3-hexafluoropropan-2-ol (molecular weight: 168.04) were introduced into a three-necked flask of capacity 2 litres which was furnished with a stirrer and a thermometer. While stirring the liquid mixture, 843.58 g of sulphuric acid were added from a dropping funnel over a period of 30 minutes. A rise in the liquid temperature was
10 observed as a result of the addition of the sulphuric acid and the liquid temperature when the addition was started was 15°C and the liquid temperature when the addition had been completed was 25°C. A dry-ice trap was fitted and the system was sealed with nitrogen and the reaction liquid was stirred while maintaining the liquid temperature at 50°C. Seven
15 hours after the start of the addition of the sulphuric acid the reaction liquid was transferred to a beaker but no loss in weight of the reaction liquid was observed. On neutralizing the reaction liquid with aqueous sodium hydroxide solution it separated into two phases and 1435.9 g of the heavier liquid was obtained using a separating funnel. This heavier liquid was
20 analyzed using gas chromatography and it was found to contain 39.87 wt% of $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ of molecular weight 348.09 which is the target compound (yield based on 1,3,5-trioxane 44%). The heavier liquid also contained unreacted 1,1,1,3,3,3-trifluoropropan-2-ol and a by-product which thought to have the structure $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}_2\text{OCH}(\text{CF}_3)_2$. This
25 by-product was included in the heavier liquid in an amount of 20.59 wt% (yield based on 1,3,5-trioxane 42%). Unreacted 1,3,5-trioxane and formaldehyde which is a related compound were not detected in either the heavier liquid or the lighter liquid. The abovementioned heavier reaction liquid obtained above was washed with water and the unreacted
30 1,1,1,3,3,3-hexafluoropropan-2-ol was removed, and then distillation was carried out at normal pressure using a liquid mixture to which diglyme had been added as the distillation raw material. As a result it was possible to

refine the $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ and about 500 g of a fraction of purity of at least 99.5 GC% were obtained

[0017]

Reference Example 2

5 The fundamental properties were measured using the refined $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ obtained in Reference Example 1. The boiling point at normal pressure was 118.2°C. The vapour pressure at 25°C at normal pressure was 1.79 kPa, the latent heat of vaporization was 129.0 kJ/kg and the solubility parameter was $6.86 (\text{cal}/\text{cm}^3)^{1/2}$. The
10 abovementioned property values are the results obtained by measuring the vapour pressure curve with the isotechniscope method. Other properties at 23°C under normal pressure and the method used for their measurement are indicated below. Density: 1617.5 kg/m³ (pycnometer method), kinematic viscosity: 0.884 cSt (capillary tube viscometer method),
15 specific dielectric constant: 4.98 (measured with the transformer bridge method, in the frequency range from 1 kHz to 1 MHz using a liquid cell), surface tension: 16.05 dyn/cm (ring method), specific heat measured using a differential scanning calorimeter: 1.186 kJ/(kg×K), solubility for water by gas chromatography: 0.01 g/100 g-H₂O.

20 [0018]

Reference Example 3

 Toxicity testing was carried out using the refined $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ obtained in Reference Example 1. The toxicity by oral administration of $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ was investigated using
25 Slc: Wistar strain rats. The test groups were dosed with 0 (solvent control group) or 2000 mg/kg as a single oral dose with five male and five female rats in a single group and then the animals were observed for a period of 14 days and the toxic signs and the LD₅₀ value were investigated. The results showed that no animal died in either the male group or the female
30 group and the general condition of the animals showed no abnormality. The change in body weight exhibited a smooth increasing transition and the amount of food ingested was normal. No changes of any note were

observed in the findings at autopsy. On the basis of the abovementioned results the LD₅₀ is in excess of 2000 mg/kg.

[0019]

Reference Example 4

5 Mutagenicity tests were carried out using the refined
(CF₃)₂CHOCH₂OCH(CF₃)₂ obtained in Reference Example 1. Whether or
not (CF₃)₂CHOCH₂OCH(CF₃)₂ had a mutation-inducing capacity was
investigated using rat typhus organisms Salmonella typhimurium TA100,
TA1535, TA98 and TA1537 and Escherichia coli WP2uvrA in accordance
10 with the "Standards for the provision of testing facilities for the investigation
of toxicity (Communication No.76 of the Ministry of Labour, dated 1st
September, 1988)" at a test facility regulated by ordinance 4 which
designates the items of toxicity to be investigated for designated chemical
substances and the testing of new chemical substances (communications
15 in respect of the environmental No.39, the drugs No. 229 and 59 basic
No.85 dated 31st March 1984, revised 18th November 1988). The results
showed that the number of reversion mutated colonies on treatment with
the substance being tested did not increase by a factor of 2 or more when
compared with a negative control with any of the bacterial strains of the
20 base pair exchange type or the frame shift type irrespective of any
activation in metabolism and neither was any responsiveness to the
amount being used observed. Hence the substance being tested can be
adjudged not to have a mutation-inducing effect under the conditions of
this test.

25 [0020]

Reference Example 5

The flash point was measured using the refined
(CF₃)₂CHOCH₂OCH(CF₃)₂ obtained in Reference Example 1, using the
flash point measurement method laid down in the test methods for
30 hazardous materials of class 4 of the fire regulation (JIS K2265-1996).
The results were such that with (CF₃)₂CHOCH₂OCH(CF₃)₂ ignition did not
occur in the temperature range from room temperature (23°C) to 80°C.
Moreover, the apparatus used was an automatic flash point tester

produced by the Rigo Co. (closed chamber type, catalogue number RFT-201) and the amount of sample supplied for testing was 50 ml. The result obtained on measuring the flash point of p-xylene (standard sample for closed chamber type flash point measurement produced by Tokyo Kasei Kogyo) before making this measurement was 27.5°C. This result conformed with the standard value for the flash point of p-xylene disclosed in JIS K2265-1996 ($27 \pm 0.5^{\circ}\text{C}$) and so it was confirmed that both the apparatus and the measurement procedure were satisfactory.

[0021]

10

Effect of the Invention

The compositions of this invention have excellent properties such as cleaning properties similar to those of freon and the chlorinated hydrocarbons and low toxicity and, moreover, by using these compositions it is possible to greatly reduce the environmental burden in terms of the destruction of the ozone layer and global warming when compared with that imposed by the freon and chlorinated hydrocarbons which have been used in the past.

15

20 **Brief Explanation of the Drawings**

Figure 1 shows the $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ concentration in the liquid phase (x1)/ the concentration of the said substance in the vapour phase (y1) curve for the vapour/liquid equilibrium state of the said substance + 1-butanol system.

25

Figure 2 shows the $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ concentration in the liquid phase (x1)/temperature (t) curve and the concentration in the vapour phase (y1)/ temperature (t) curve for the vapour/liquid equilibrium state of the said substance + 1-butanol system.

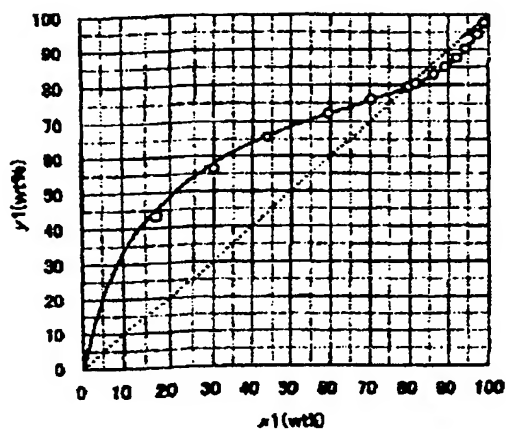
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Figure 3 shows the $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ concentration in the liquid phase (x1)/ the concentration of the said substance in the vapour phase (y1) curve for the vapour/liquid equilibrium state of the said substance + 2-butanol system.

Figure 4 shows the $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2$ concentration in the liquid phase (x_1)/temperature (t) curve and the concentration in the vapour phase (y_1)/ temperature (t) curve for the vapour/liquid equilibrium state of the said substance + 2-butanol system.

5

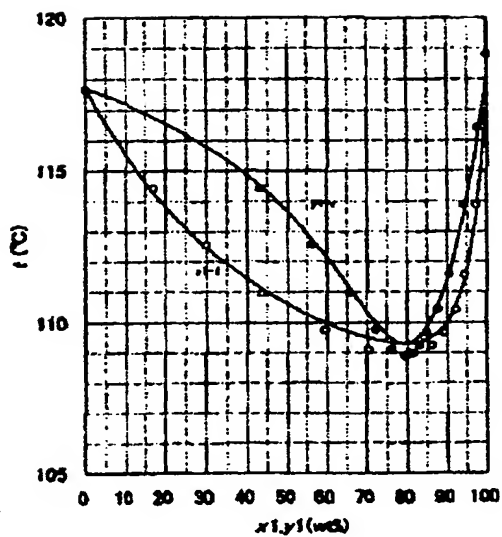
Figure 1



Vapour/Liquid Equilibrium Curve (x-y)
 $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2 + 1$ Butanol

10

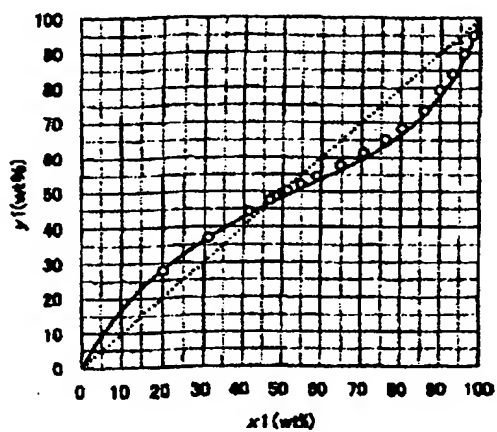
- 17 -
Figure 2



Vapour/Liquid Equilibrium Curves (x,y - t)
 $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2 + 1\text{-Butanol}$

5

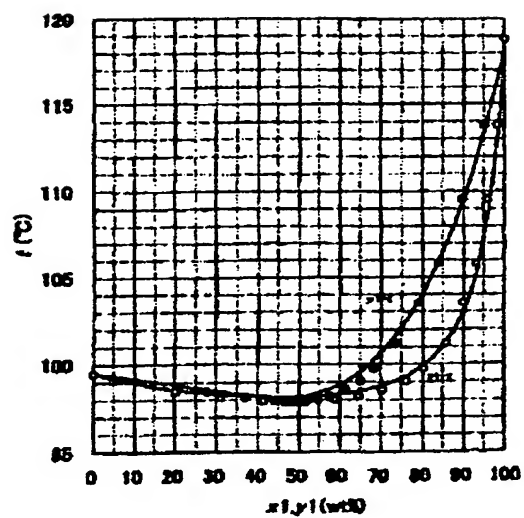
Figure 3



Vapour/Liquid Equilibrium Curve (x-y)
 $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2 + 2 \text{ Butanol}$

10

- 18 -
Figure 4



Vapour/Liquid Equilibrium Curves (x,y - t)
 $(\text{CF}_3)_2\text{CHOCH}_2\text{OCH}(\text{CF}_3)_2 + 2\text{-Butanol}$